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MOISTURE CORRECTIONS IN NEUTRON COINCIDENCE COUNTING OF PuO_2

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ABSTRACT

Passive neutron coincidence counting is capable of 1% assay accuracy for pure, well-characterized PuO_2 samples that contain plutonium masses from a few tens of grams to several kilograms. Moisture in the sample can significantly bias the assay high by changing the (α,n) neutron production, the sample multiplication, and the detection efficiency. Monte Carlo calculations and an analytical model of coincidence counting have been used to quantify the individual and cumulative effects of moisture biases for two PuO_2 sample sizes and a range of moisture levels from 0 to 9 wt%. Results of the calculations suggest a simple correction procedure for moisture bias that is effective from 0 to 3 wt% H_2O. The procedure requires that the moisture level in the sample be known before the coincidence measurement.

I. INTRODUCTION

Neutron coincidence counters are used routinely in U.S. Department of Energy (DOE) and international facilities for quick verification measurements of a wide variety of uranium- and plutonium-bearing materials. Typically, active interrogation is the preferred technique for uranium and passive counting for plutonium. The most frequently used passive neutron counter is the HLNC-II,[1] which is the standard instrument for ^{240}Pu (effective) verifications made by the International Atomic Energy Agency and EURATOM-Luxembourg inspectorates. The ^{240}Pu (effective) determination is combined with a plutonium-isotopic measurement to yield an independent verification of sample plutonium mass.

Assuming a set of pure PuO_2 samples with known and uniform plutonium isotopics and uniform bulk density, the plutonium mass of verification samples from the set can be measured to better than 1% accuracy in 5 min of counting using only the uncorrected real coincidence rate. An algorithm that corrects for sample/detector neutron multiplication effects[2] yields <1% accuracy for pure PuO_2 samples with known but variable plutonium isotopics and variable bulk density.

II. PROBLEM STATEMENT

Residual moisture can be present in PuO_2 samples that must be verified using neutron coincidence counting. Moisture affects the uncorrected real coincidence count rate in three ways. First, the number of neutrons produced by (α,n) reactions in the sample is increased. Second, the number of induced-fission neutrons (multiplication) is increased because hydrogen lowers the neutron energy. Third, the detection efficiency is increased because of the lowered neutron energy. The Sellafield (United Kingdom) exercise of January 1985 identified three samples that showed positive bias in both uncorrected and multiplication-corrected coincidence rates.[3] The biases were ascribed to several possible causes, including H_2O contamination.

III. CALCULATIONAL METHOD

A hybrid Monte Carlo/analytical model[4] has been used to calculate coincidence count rates for a 3.03-kg PuO_2 sample (Sellafield sample #17) and a 1.0-kg sample (Los Alamos sample LA0261C11) measured in an HLNC-II. The H_2O content of the samples was varied from 0 to 9 wt%. The HLNC-II/sample geometry and materials were modeled from design drawings, Los Alamos destructive analyses, and information from Ref. 3.

For the Sellafield sample, the bulk density used for the dry PuO_2 was 2.642 g/cm^3. This value was obtained from results of parametric multiplication calculations made by P. Rinard.[5] The dry-material bulk density used for the Los Alamos PuO_2 sample was 2.502 g/cm^3. This value was obtained from the net weight and a radiograph to obtain the fill height. Table I defines characteristics of the two samples used for the calculations. Note the Sellafield α is more than a factor of 2 larger than the Los Alamos α.

TABLE I

SAMPLE CHARACTERISTICS

Characteristic	Sample	
	Sellafield 017 (as of 1/85)	Los Alamos LA0261C11 (as of 4/86)
Pu(g)	2072	875.6
^{238}Pu (wt%)	0.915	0.059
^{239}Pu (wt%)	67.58	82.07
^{240}Pu (wt%)	23.32	16.36
^{241}Pu (wt%)	4.59	1.17
^{242}Pu (wt%)	3.63	0.341
$^{241}Am/Pu$ (wt%)	3.55	0.231
^{240}Pu-eff (g)	636	149.2
$^{239+241}Pu$ (g)	1927	728.9
α[a]	0.855	0.392
$\rho(g/cm^3)$[b]	2.642	2.502

[a] α--the ratio of (α,n) to spontaneous fission neutrons emitted in the sample. Values listed are for dry, pure PuO_2; that is,

$$\alpha = \frac{134\, f_{238} + 0.381\, f_{239} + 1.41\, f_{240} + 0.013\, f_{241} + 0.02\, f_{242} + 26.9\, f_{241Am}}{10.22\,(2.43\, f_{238} + f_{240} + 1.69\, f_{242})}$$

where the f_i's are weight percents of plutonium isotopes and ^{241}Am.

[b] ρ--dry-sample bulk density used in calculation.

IV. CALCULATIONAL RESULTS

Results of the Sellafield sample calculations are shown in Table II. Real coincidence count rates before multiplication correction (BMC) were calculated for samples with 0, 1, 3, 5, 7 and 9 wt% H_2O using the formalism of Ref. 4. Water was assumed to increase the sample density, not its volume. The BMC coincidence count rates increase with increasing moisture, approaching +30% compared with the dry count rate for the 9 wt% H_2O case. However, for fired PuO$_2$ powder, 2-3 wt% H_2O appears to be a practical upper limit for the majority of cases of interest. This observation is made based on destructive analyses of selected moist samples in European and DOE facilities. As Table II shows, the BMC moisture bias at 3 wt% H_2O from all effects is +8%. The efficiency (ϵ) bias component and the α bias component are both -2.2% whereas the induced-fission multiplication (M) component is +3.4%. Note these effects do not add to the total because the equation for the reals rate is nonlinear in ϵ and M and contains products of all three variables. Individual bias components were determined by substituting appropriate calculated values of ϵ, M, and α into the exact expression for the coincidence count rate (see Ref. 4). For example, to obtain the 3.4% M bias, the dry-sample values for ϵ and α and the wet (3 wt% H_2O) value of M were inserted into the equation. Figure 1 is a plot of the wet-to-dry ratio of the BMC or uncorrected reals rate vs moisture content for the Sellafield and Los Alamos samples. The two point sets represent the biases from all effects.

Table II also shows moisture biases in after-multiplication-correction (AMC) coincidence count rates. These corrected rates were obtained exactly as if the calculated coincidence rates, R, and totals rates, T, had been measured and then processed with the multiplication-correction (MC) algorithm.[4] The algorithm requires the parameters ρ_0 and K (along with the measured values of R and T), where

$$\rho_0 = \frac{R_0}{T_0}(1 + \alpha_0) = \frac{\epsilon_0 f_0}{2}\,\frac{\overline{\nu(\nu-1)}_s}{\bar{\nu}_s} \qquad (1)$$

and

$$K = \frac{\overline{\nu(\nu-1)}_I}{\bar{\nu}_I - 1} \Bigg/ \frac{\overline{\nu(\nu-1)}_s}{\bar{\nu}_s} \qquad (2)$$

TABLE II

PERCENT BIASES IN REAL COINCIDENCE RATES
CAUSED BY MOISTURE (0-9 wt% H_2O)
SELLAFIELD SAMPLE #17 (3.03 kg PuO_2)

$$\frac{R_{wet} - R_{dry}}{R_{dry}} \times 100$$

	Before Multiplication Correction				After Multiplication Correction			
wt% H_2O	All Effects	ϵ^a	M^b	α^c	All Effects	ϵ	M	α
0	-	-	-	-	-	-	-	-
1	0.9	-0.8	0.9	0.8	2.2	-0.2	-	2.5
3	8.0	2.2	3.4	2.2	7.7	0.9	-	6.8
5	14.9	5.1	5.4	3.5	12.9	2.0	-	10.8
7	25.4	10.6	7.9	4.7	18.8	3.7	-	14.6
9	29.5	9.1	11.6	5.8	21.7	3.2	-	17.9

[a] ϵ is the bias resulting from the detection efficiency change with moisture
[b] M is the bias resulting from the multiplication (induced-fission) change with moisture
[c] α is the bias resulting from the change in (α,n) reactions with moisture.

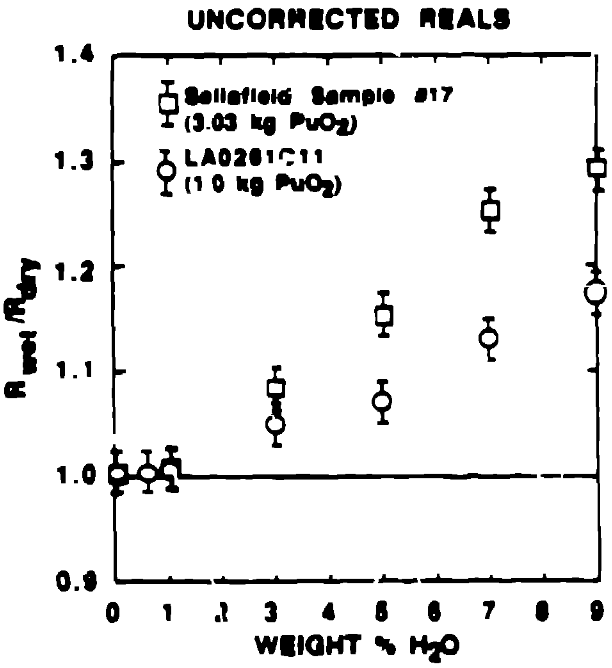

Fig. 1. Calculated HLNC-II coincidence response ratio before multiplication correction as a function of moisture content in PuO_2.

The parameter ρ_0 pertains to a nonmultiplying sample and contains detection efficiency, ϵ_0, fraction of pulses counted in the coincidence gate, f_0, and the first and second reduced moments of the spontaneous-fission multiplicity distribution. The parameter K contains first and second reduced moments of both spontaneous and induced fission multiplicity distributions. Values of ρ_0 = 0.1074 and K = 2.131 were calculated using Monte Carlo averages of ϵ_0, f_0, $\bar{\nu}^I$ and $\overline{\nu(\nu-1)}^I$ for the range of Los Alamos sample masses (see Ref. 4). Values of $\bar{\nu}^S$ = 2.156 and $\overline{\nu(\nu-1)}^S$ = 3.823 were taken from Ref. 7.

As indicated in Table II, the moisture bias from all effects for the corrected reals rates (AMC) increases with increasing moisture content approaching 22% for 9 wt% H_2O. This value is almost three-fourths that of the BMC bias. For the 3 wt% H_2O case, the AMC bias is -7.7% from all effects. This value is nearly equal to the corresponding BMC bias. Table II also shows that the ϵ component of bias for 3 wt% H_2O is -0.9%, the M component is zero, and the α component is -6.8%. These AMC bias components were obtained using a procedure extending that used for the BMC components. Appropriate values of ϵ, M, and α were substituted into the exact expressions for R and T. These were then substituted into the MC algorithm (along with the appropriate value for ρ_0) to yield the AMC value for R (R_c). As an illustration, to obtain the -0.9% ϵ bias, wet-sample values of ϵ, M, and 1 were used to calculate R and T. These were used with the dry-sample value of ρ_0 to yield R_c from the MC algorithm. This procedure isolates the bias resulting from the fact that ε has increased because of moisture and ρ_0 contains the wrong dry-sample values for efficiency (ε_0) and gate fraction (f_0). The multiplication (M) bias component is removed by the definition of the MC algorithm. That is, if the moist values for α and ρ_0 are used, the algorithm automatically corrects for multiplication effects, even if produced by moisture.

Table II shows that by far the largest AMC bias component arises from use of dry-sample α in the MC algorithm. Note also that the AMC α bias is roughly three times the BMC α bias component. If the moisture contamination range 0-3% is considered, the (α,n) bias after multiplication correction is the only significant component.

Figure 2 is a plot of the wet-to-dry ratio of the AMC or multiplication-corrected coincidence count rates vs moisture content for the Sellafield and Los Alamos samples. The upper point sets describe the biases from all effects and the lower point sets are the detector efficiency (ε) biases only. Note the bias effects are uniformly smaller for the sample with the smaller mass and the smaller α.

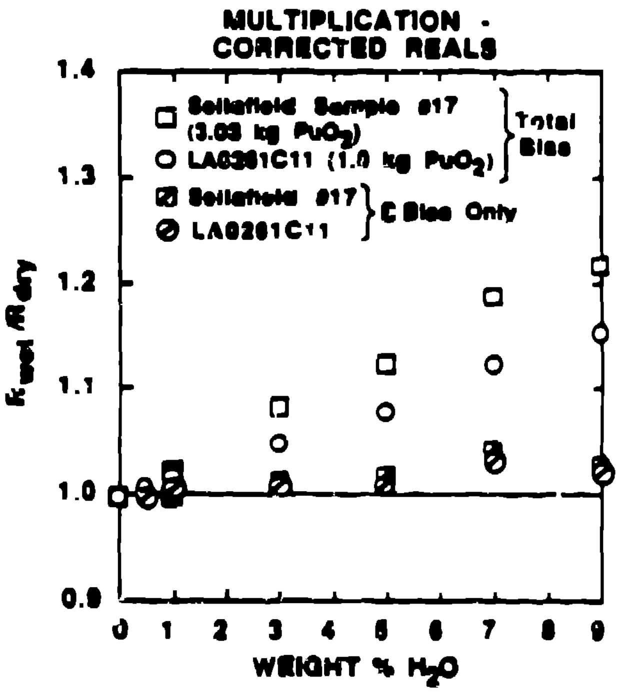

Fig. 2. Calculated HLNC-II coincidence response ratio after multiplication corrections as a function of moisture content in PuO_2.

Table III shows the same results as Table II (Sellafield sample) for the Los Alamos sample. The BMC and AMC biases from all effects are smaller for the Los Alamos sample compared with those for the Sellafield sample. The BMC bias components for ε and M are smaller because the Los Alamos sample contains approximately one-third the H_2O mass when compared with the Sellafield sample at the same wt% H_2O. The extra H_2O in the Sellafield sample produces a softer neutron spectrum, which increases the ε and M bias components. The Sellafield dry-sample α is 2.18 times that of the Los Alamos sample. This difference increases the bias arising from using a dry-sample α to analyze a wet-sample measurement. This result is reflected in the BMC and AMC bias components being uniformly larger for the Sellafield sample than for the Los Alamos sample.

V. MOISTURE-CORRECTION PROCEDURE

In the algorithm that removes multiplication (induced-fission) contributions to the measured coincidence count rate, the (α,n) neutron production term appears through the parameter α, which is defined as the number of (α,n) neutrons produced in the sample divided by the number of spontaneous-fission neutrons produced. If we define α as the appropriate ratio for a wet sample and α_0 as that for a dry one, data from SOURCES code[8] calculations have been fitted extremely well with the quadratic

$$\alpha/\alpha_0 = a_2x^2 + a_1x + a_0 , \qquad (3)$$

where x = wt% H_2O,
$a_2 = -0.0007663$,
$a_1 = 0.04387$, and
$a_0 = 1.0005$.

This expression is independent of sample plutonium-isotopic content.

Results of the Monte Carlo and SOURCES code calculations suggest the following procedure for removing the effects of moisture from neutron coincidence counting:

(1) Determine the moisture content of the sample. One may take the operator's process value or, if an independent verification is required, one may use such approaches as nuclear magnetic resonance, dual-ring spectral index measurements using a neutron coincidence counter, and bare ^3He detector measurements.

(2) Calculate α_0 for the dry sample using the normal procedure and determine α from the known moisture percentage and Eq. (3).

TABLE III

PERCENT BIASES IN REAL COINCIDENCE RATES
CAUSED BY MOISTURE (0-9 wt% H_2O)
LOS ALAMOS SAMPLE LA0261C11 (1 kg PuO_2)

$$\frac{R_{wet} - R_{dry}}{R_{dry}} \times 100$$

wt% H_2O	Before Multiplication Correction				After Multiplication Correction			
	All Effects	ϵ^a	M^b	α^c	All Effects	ϵ^a	M^b	α^c
0	-	-	-	-	-	-	-	-
0.58	0.1	-0.2	0.2	0.2	0.8	-0.1	-	0.9
1	1.0	0.3	0.4	0.3	1.7	0.1	-	1.6
3	5.0	2.8	1.3	0.7	5.2	0.8	-	4.2
5	6.7	3.0	2.4	1.2	7.9	1.0	-	6.8
7	13.3	7.8	3.3	1.6	12.3	2.7	-	9.3
9	17.5	8.9	5.6	2.0	14.8	3.0	-	11.4

[a] ϵ is the bias resulting from the detection efficiency change with moisture.
[b] M is the bias resulting from the multiplication (induced-fission) change with moisture.
[c] α is the bias resulting from the change in (α,n) reactions with moisture.

(3) Measure the sample in an HLNC-II and apply the multiplication correction using the wet value of α as determined from step (2).

The results of Tables I and II show that this procedure should yield <1% bias arising from detector efficiency changes for moisture values <1 wt% H_2O. If the HLNC-II moderator were increased, this small bias would also disappear.

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